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Summary of the Environmental Radiological Monitoring Program at Lawrence Livermore National Laboratory

P. E. Althouse

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Disclaimer

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Paris E. Althouse
Environmental Protection Department, Lawrence Livermore National Laboratory
PO Box 808, Livermore, CA 94550 USA

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LLNL Mission

Lawrence Livermore National Laboratory (LLNL) is a premier, US Department of Energy, applied-science, national security laboratory, whose primary mission is to ensure that the nation's nuclear weapons remain safe, secure, and reliable and to prevent the spread and use of nuclear weapons worldwide. This mission enables our programs in advanced defense technologies, energy, environment, biosciences, and basic science to apply LLNL's unique capabilities, and to enhance the competencies needed for our national security mission. The Laboratory serves as a resource to the US government and as a partner with industry, academia, and international communities.

With local, state, and federal interests, LLNL is committed to providing responsible stewardship of the environmental resources in our care. LLNL has developed a rigorous radiological monitoring program that will be summarized in this paper.

LLNL Setting

LLNL operates two sites: the Livermore main site (or Site 200) and a nearby experimental test site (Site 300) (Figure 1). The Livermore main site is approximately 260 ha located near the San Francisco Bay in Northern California, and is situated in the town of Livermore, which has a population of 74,000. There are residential neighborhoods within 100 m of the main site, and more than 45% of LLNL employees live in the Livermore Valley. Site 300 is a remote testing facility in the hills 24 kilometers east of the main site. Radiological monitoring activities are conducted at each site.

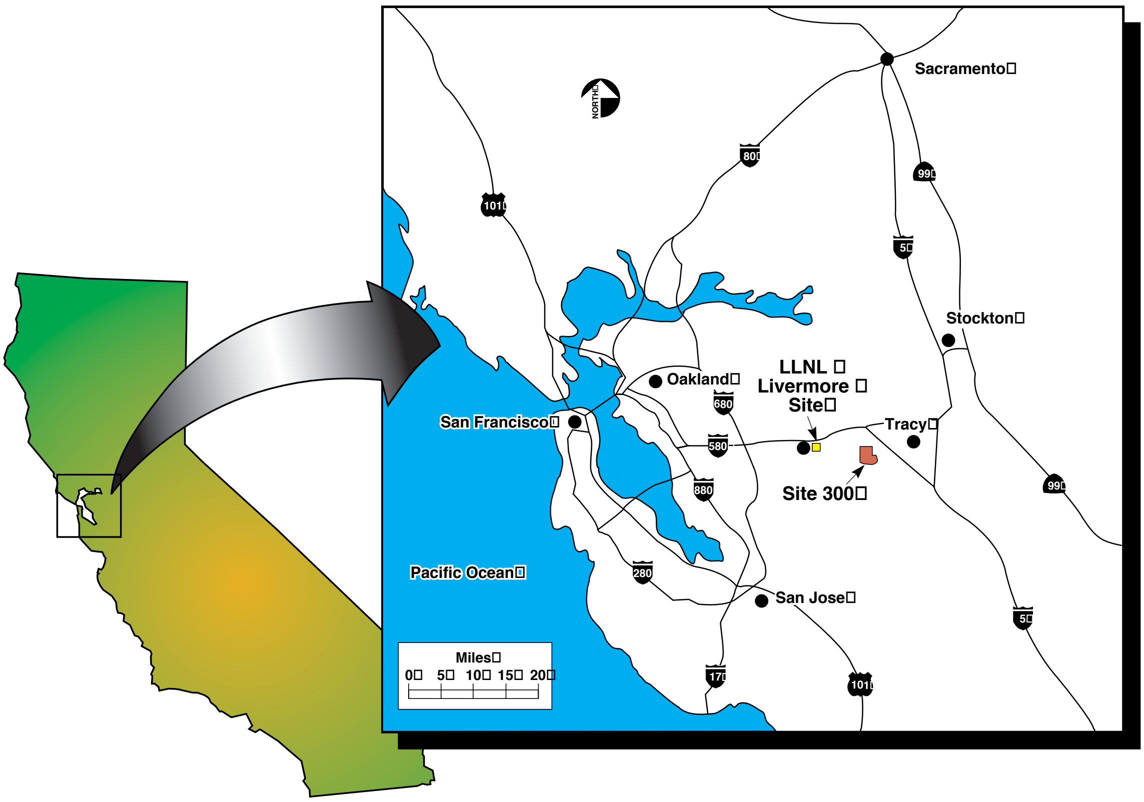

Figure 1. LLNL and Site 300 Location

Environmental Protection Department Mission

LLNL has an Environmental Protection Department (EPD) that is responsible for environmental monitoring, environmental regulatory compliance, environmental restoration, and hazardous waste management at both sites. EPD provides assistance to LLNL programs through support activities such as:

- Conducting environmental evaluations and addressing requirements under federal, state, and local laws and regulations.
- Identifying and developing methods to monitor, prevent, reduce, and clean up air emissions, wastewater discharges, and hazardous wastes.
- Obtaining permits or exemptions for air, water, and hazardous waste activities.
- Ensuring environmental compliance through environmental monitoring, risk assessment, and analysis of LLNL activities. Specifically, EPD evaluates the impact of ongoing LLNL operations on the surrounding environment by sample collection, analysis, data reduction, and simulation modeling.
- Developing and conducting cost-effective environmental restoration and remediation.

• Designing and applying appropriate, cost-effective treatment technologies to manage hazardous and nonhazardous waste streams.

• Developing and implementing waste minimization and pollution abatement strategies.

• Coordinating Laboratory-wide decontamination and decommissioning activities.

EPD publishes a Site Annual Environmental Report (SAER) each year detailing the results of all environmental monitoring performed, both radiological and non-radiological (Sanchez et al., 2002). In addition, every three years, an Environmental Monitoring Plan (EMP) is updated (Woods, ed., 2002) which describes all environmental monitoring performed by LLNL (including non-radiological monitoring activities). Each chapter in the EMP contains the following components:

• Introduction to the medium being described: example air, water, soils, etc.

• Outline of regulatory drivers and requirements

• Evaluation of potential site sources and contaminants

• Description of the extent and frequency of monitoring

• Explanation of sampling locations

• Description of collection and analytical methods

• Discussion of action levels and reporting requirements

The environmental monitoring program has two major components. First, the program directly monitors effluents, such as stack emissions, as well as storm drain and sanitary sewer discharges. Second, the program conducts surveillance monitoring of all environmental media that could be impacted by LLNL, including air, surface water, groundwater, rainwater, surface runoff, wastewater and sewage, vegetation and foodstuffs, soils and sediments, and direct radiation. This existing program involves a staff of over 50 Laboratory scientists and technicians as well as contractors; the collection of more than 24,000 samples from a variety of environmental media; and the performance of more than 260,000 analyses per year.

The EMP for radiological monitoring activities includes many different facets of "monitoring," from collection of samples to modeling LLNL's impact for regulatory compliance and assessments (Figure 2).

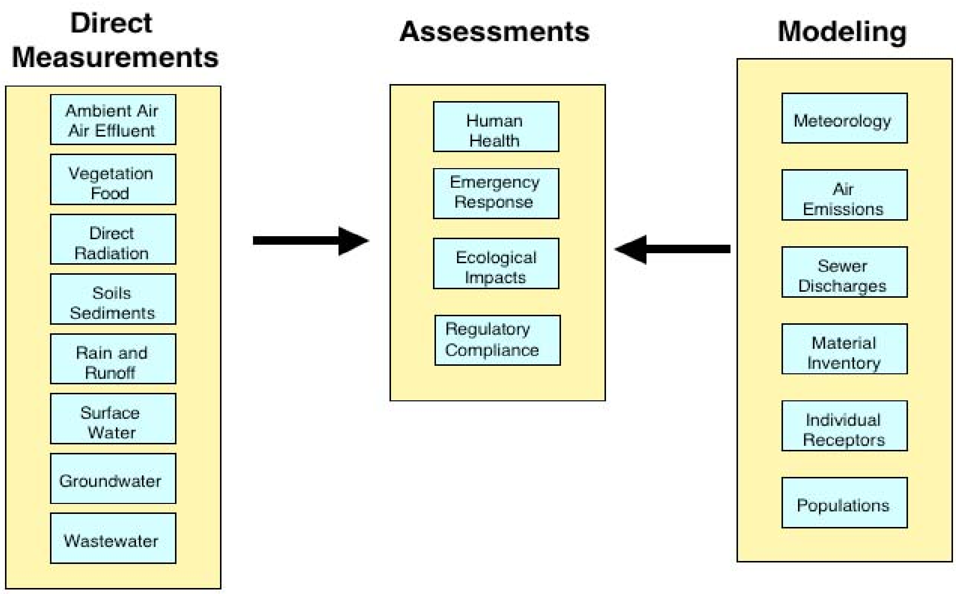

Figure 2. General facets of monitoring

EMP Sample Collection and Methodology

<u>Air Monitoring</u>

Air-Effluent

The primary purpose of LLNL's air effluent sampling program is to measure radiological emissions at the point of release (at the stack). LLNL operates 72 continuous samplers in seven facilities at the Livermore site and one continuous sampler in one facility at Site 300. Samples of stack emissions are collected on filters, ionization chambers, and real-time continuous air monitoring systems (CAMS) and are analyzed for gross alpha and gross beta particles, and tritium. Several of these systems have real-time alarm capabilities and provide immediate data.

Air-Surveillance

Air surveillance differs from air effluent in that samples are collected and analyzed in a laboratory; therefore, data are not immediately available. To monitor radiological particulates and airborne tritium vapors, LLNL maintains 27 high volume air particulate samplers (collected weekly) and 17 vapor collection samplers (collected bi-weekly). All surveillance-sampling units are positioned to ensure reasonable probability that any significant concentration of radiological effluents of concern from LLNL operations will be detected. Samples for particulate are collected on cellulose filters, while tritium vapors are collected in silica gel.

Analyses for particulate samples include gross alpha and gross beta and gross screening measurements for isotopic gamma emitters. Particulate samples are then digested and portions of the filter papers are analyzed for isotopic determination of plutonium (by alpha spectrometry) and uranium (by inductively coupled plasma mass spectrometry). Air vapor samples are analyzed using liquid scintillation after extracting the vapor/moisture from silica gel using a freeze dry method.

Real-time Radiation in Air

For real-time radiological area monitoring and to provide information in the event of accidental release from non-monitored sources, LLNL maintains 16 perimeter geiger-mueller sensors. These real-time sensors are linked to the LLNL emergency operation center and can be accessed during radiological emergency situations (Bertoldo, 2004).

Direct Radiation by Thermoluminescent Dosimeters (TLD)

Environmental surveillance and effluent monitoring for radionuclides, followed by pathway and dose assessments is one method to determine LLNL-induced radiological impacts, but for completeness, direct radiation impacts must also be evaluated. Penetrating radiation cannot be measured by collection of material on filters nor chemically trapped; it is collected by trapping the penetrating gamma radiation in the crystal lattice of solid-state devices known as thermoluminescent dosimeters (TLDs.)

The primary purpose of direct radiation monitoring is to measure radiation doses and evaluate the dose received by the public, if any, from direct gamma radiation originating at LLNL. TLDs are placed around the perimeter of LLNL, (co-located with the real-time monitoring geiger-mueller sensors), at Site 300, and at off-site locations for a total of 65 locations. TLDs are read in a three phase process. TLDs measure exposure as absorbed dose (in milliroentgen; mR).

Sewer Discharge and Wastewater Monitoring

The research and development activities at LLNL require the use of hazardous and radioactive materials; if significant concentrations of these materials were inadvertently discharged to the sanitary sewer, they could seriously impact the local water reclamation plant operations and potentially degrade the quality of water resources. To limit potential discharge, LLNL has developed administrative and engineering controls. Administrative measures include implementation of internal discharge limits, training of materials handlers, control and tracking of certain materials, drain labeling, and inspection and review of facilities and operations. Engineering controls include isolating specific operations from sanitary connections and collecting industrial wastewater from entire facilities in retention tanks.

The sewer monitoring program supports the discharge control effort in three ways. First, it defines criteria for the acquisition and analysis of retention tank samples. Second, sampling and analysis of the sewage actually released enable EPD to assess the effectiveness of the control program in maintaining pollutant levels below discharge limits. Third, LLNL continuously monitors in real time for those pollutants with the greatest potential to adversely impact the public welfare.

There are currently 45 in-service wastewater retention tank systems at LLNL, including those at Site 300. Sampling frequency for retention tanks is determined by operations. Primary radiological wastewater concerns are: tritium, carbon-14, potassium-40, uranium, plutonium, and americium. When full, retention tanks are analyzed for gross alpha and beta activity. If the results are higher than an established action limit, isotopic determination is performed on the samples.

Surface Water Monitoring

LLNL monitors the radioactivity in surface water at the swimming pool, an on-site drainage basin, local rainfall, the domestic water supply, reservoirs, ponds, and storm water. Samples are analyzed for gross alpha, gross beta, plutonium, uranium, and tritium. Storm water is monitored before coming on site and after leaving to determine if any radioactivity was introduced by LLNL.

Groundwater

LLNL's groundwater monitoring program has both surveillance and compliance objectives. Surveillance monitoring requires continual radiological assessment of all LLNL activities for their potential to release contaminants to groundwater. Surveillance monitoring programs are in place at both the Livermore site and Site 300, and at offsite locations. Compliance monitoring, currently conducted at Site 300 only, detects potential releases to groundwater of radiological constituents from landfill pits, surface water impoundments, and sewage ponds. Ground water samples are collected and analyzed for gross alpha and beta, tritium, strontium-90, radium, and uranium radioisotopes. Each perimeter surveillance monitoring well is also sampled and analyzed, annually at a minimum, for plutonium-238, plutonium-239+240, thorium-228, thorium-230, thorium-232, and americium-241 by alpha spectroscopy. Cesium-137, cobalt-60, and potassium-40 are measured by gamma spectroscopy.

Soil and Sediment Monitoring

Surface soils are collected annually from the top 5 centimeters of soil while sediment samples are collected at depths of 5, 15, 45, and 60cm. Soil sampling and analysis is performed to determine if there is measurable long-term buildup of radionuclides in the terrestrial environment. Surface soil sampling locations are determined by proximity to LLNL sites and the potential for being affected by LLNL operations from wind deposition of contaminants. Other selection criteria include background locations with geologically similar substrates as those near each site (but unlikely to be affected by LLNL operations), areas of known or suspected LLNL-induced contamination, and proximity to an air sampling location to enable analysis of re-suspension. To satisfy these requirements, surface samples are collected at 32 locations.

The radiological analytes of greatest interest for the Livermore site are plutonium and tritium. All soil and sediment samples are analyzed for plutonium and gamma-emitting nuclides; sediment samples are also analyzed for tritium because these samples are taken in locations that channel water on the site.

The radiological analytes of interest at Site 300 are the isotopes of uranium, especially uranium-238 and uranium-235, and the ratio of these values in a given sample. Depleted uranium (i.e., natural uranium depleted of much of the uranium-235) has historically been, and is currently used in experimental tests at Site 300.

Analytical methods used for soil include plutonium-238 and plutonium-239+240 by alpha spectroscopy following acid leaching, gamma scans using a high purity germanium detector, and tritium by liquid scintillation following freeze-dry extraction of the soil moisture.

Vegetation and Foodstuff

Sampling and analysis of vegetation and foodstuff can provide information about the presence and movement of radionuclides released to the environment. At LLNL, vegetation and wine are sampled. Wine is sampled because it is the most important agricultural product in the Livermore Valley, representing an approximately $140-million annual industry. Concentrations of radionuclides in vegetation can be used to estimate concentrations in edible plant and animal products and dose to humans from ingestion of a normal diet. Historically, milk and honey samples were collected directly from owners of local cows or goats and from beekeepers.

Tritium is the only nuclide released from LLNL that can be detected in vegetation; therefore it is the only radioisotope analyzed. Samples are collected quarterly from only the green, leafy material of grass and other vegetation and collected from on-site, off-site, upwind and downwind locations.. The vegetation is stored frozen until analysis, at which time water is extracted from the samples by freeze-drying techniques. The samples are then analyzed for tritium by liquid scintillation counting.

The annual wine sampling is an extremely sensitive issue because of the potential economic, political, and public relations impacts of the data. Wine is analyzed for tritium content by area: Livermore Valley, other California wine regions, and from Europe (France, Germany, and Italy). European wines were initially chosen for evaluation because Europe is a significant wine-growing region with historically high tritium content in wine from locations near nuclear power plants. California wines from regions other than the Livermore Valley serve as natural background samples for comparative purposes. Because geographic variability of the wines is addressed, wines selected from a number of regions roughly represent the tritium exposure received by the wine-drinking public. Wine samples are analyzed for tritium content by helium-3 mass spectrometry.

Dose and Risk Assessment

The ultimate purpose for all sampling and monitoring is to determine potential radiological doses to individuals and populations caused by releases of radionuclides to the environment from LLNL. LLNL determines the degree of public exposure from a combination of computer modeling and physical measurements as described earlier. LLNL's primary calculational tool for estimating dose and risk from routine operations is the computer code CAP88-PC. The code computes dose and risk to both individuals and collective populations from chronic or long-term radiological emissions to air. Other radiological air-dispersion modeling codes (HOTSPOT and NEWTRIT) are used at LLNL as needed to address unplanned releases or short-term releases from experiments or operations. Modeled doses tend to be conservative and therefore higher that those doses measured at the sampling locations (Figure 3).

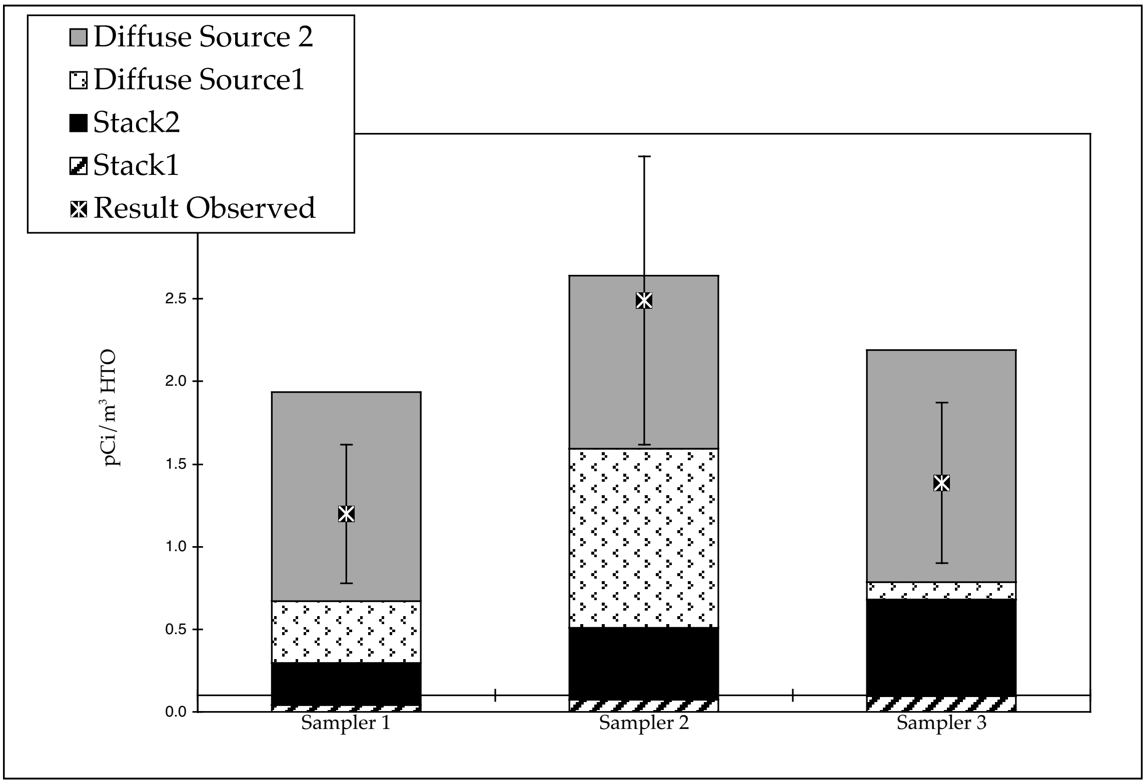

Figure 3. Direct measurement compared to modeled stack output

Quality Assurance Program Special Considerations

When developing a radiological monitoring plan such as the program deployed by LLNL, there are a number of special considerations that each monitoring network should consider and implement in the program.

A formal quality assurance (QA) program must be developed. A QA program is essential and must be considered at every level, including the analytical practices and methods. The quality assurance program should include the following sections:

- Providing training, formal standard operating procedures, and documentation of the qualifications of staff
- Quality improvement through use of non-conformance reports, audits and self assesments.
- Comprehensive and legal record managament practices
- Safety work processes; safety plans, instructions, handling, storage and shipping procedures
- Regulation identification and reporting criteria

Conclusion

In conclusion, LLNL has a rigorous and comprehensive radiological monitoring program consisting of sampling a wide array of media for number of different radioisotopes. The current techniques LLNL uses for radiological environmental monitoring are very sensitive, allowing detection of extremely low levels of contaminants. The combination of effluent and surveillance monitoring, together with computer modeling shows that radiological doses to the public caused by LLNL operations are less than 1% of regulatory standards and about 2500 times smaller than the doses received from natural background radiation. The results annually show extremely low contaminant levels, reflecting the responsiveness of the Laboratory in controlling radiological pollutants.

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